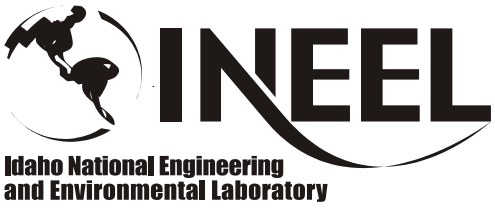
INEEL/CON-04-01571
PREPRINT



Nuclear Isotopic Dilution of Highly Enriched Uranium-235 and Uranium-233 By Dry Blending Via the RM-2 Mill Technology

R. N. Henry (INEEL)
N. A. Chipman (INEEL)
R. K. Rajamani (University of Utah)
S. Latchireddi (University of Utah)
V. Devrani (University of Utah)
H. Sethi (University of Utah)
J. L. Malhotra (NETL)

February 29, 2004 – March 4, 2004

Waste Management 2004 Symposia

NUCLEAR ISOTOPIC DILUTION OF HIGHLY ENRICHED URANIUM-235 AND URANIUM-233 BY DRY BLENDING VIA THE RM-2 MILL TECHNOLOGY

R. N. Henry, N. A. Chipman
Idaho National Engineering and Environmental Laboratory
PO Box 1625
Idaho Falls, ID 83415

R. K. Rajamani, S. Latchireddi
V. Devrani, H. Sethi
University of Utah
1471 Federal Way
Salt Lake City, UT 84102

J. L. Malhotra
National Energy Technology Laboratory
3619 Collins Ferry Road
Morgantown, WV 26507

ABSTRACT

The United States Department of Energy has initiated numerous activities to identify strategies to disposition various excess fissile materials. Two such materials are the off-specification highly enriched uranium-235 oxide powder and the uranium-233 contained in unirradiated nuclear fuel both currently stored at the Idaho National Engineering and Environmental Laboratory. This report describes the development of a technology that could dilute these materials to levels categorized as low-enriched uranium, or further dilute the materials to a level categorized as waste. This dilution technology opens additional pathways for the disposition of these excess fissile materials as existing processing infrastructure continues to be retired.

INTRODUCTION

The United States Department of Energy (DOE) has initiated numerous activities to focus on identifying material management strategies to disposition various excess fissile materials. In particular the Idaho National Engineering and Environmental Laboratory (INEEL) has stored 1,700 kg of off-specification, highly-enriched uranium (HEU) at the Idaho Nuclear Technology Energy Center (INTEC) vault facility (CPP-651). This HEU is in the form of UO_3 powder. Currently, the proposed strategies for dispositioning this material are (a) aqueous dissolution and down blending to low-enriched uranium (LEU) via facilities at the Savannah River Site (SRS) followed by shipment of the liquid LEU to Nuclear Fuel Services (NFS) for fabrication into light water reactor (LWR) fuel for the Tennessee Valley Authority (TVA) reactors, and (b) dilution of the HEU to 0.9 wt% U-

235 for discard as a waste stream that would no longer have a criticality or proliferation risks without being processed through some type of enrichment scheme.

The other fissile material of concern is U-233 currently stored at INEEL in various nuclear fuel forms. The majority of the 350 kg inventory is in the form of unirradiated light water breeder reactor (LWBR) fuel stored in a secure facility at INTEC (CPP-749) and unirradiated fuel components stored at the Radioactive Waste Management Complex (RWMC). Additional inventories of U-233 in the form of spent nuclear fuel also reside at INEEL but are not addressed in this study. Proposed strategies for dispositioning this unirradiated material include (a) direct disposal at a repository, (b) aqueous dissolution followed by isotopic dilution with natural uranium (NU) or depleted uranium (DU) to generate a new reactor fuel form or a durable form for disposal, (c) chemical processing to salvage radioisotopes for potential medical uses, and blending of the remaining fissile material into a new disposal form, and (d) physical dilution of the U-233 to about 0.6% with DU or NU for discard as a waste stream. This waste stream would no longer have criticality or proliferation risks currently associated with this material.

Isotopic dilution of HEU to typically less than 20 wt% U-235 by dry blending is the key to solving the dispositioning issue (i.e., proliferation) posed by HEU stored at INEEL. Dilution reduces storage costs because the uranium no longer requires Category I and/or II safeguards and security (S&S) measures, promotes as-low-as-reasonably-achievable (ALARA) radiation exposure philosophy for shippers and receivers because the source term has been reduced and normalized, and most importantly could decouple this material from the mission-completion schedule of the SRS processing facilities by allowing it to be processed at other HEU dilution sites.

Isotopic dilution of U-233 provides similar cost and risk benefits described for HEU. Because of criticality and proliferation concerns, it is unlikely that this material can be dispositioned without reducing its enrichment below the weapons grade threshold (10 to 12% for U-233). Aqueous dissolution and dilution offers some advantages however, anticipated costs are very high and sufficient processing may not be available in existing facilities. The partitioning of medical radioisotopes from this material remains open, but the path forward is poorly defined. Thus isotopic dilution by grinding and blending with DU or NU provides a path forward for disposal of the U-233 inventory.

The RM-2 mill technology was previously selected to study isotopic dilution by dry milling and blending. The RM-2 mill (see Figure 1), developed at the University of Utah (U.S. patent #6086242 July 2000), is a technology developed and successfully tested for producing ultra-fine particles by dry grinding. Grinding action in theRM-2 mill produces a two million-fold increase in the number of particles being blended in a centrifugal field. The concept of achieving complete and adequate blending and mixing (i.e., no methods were identified to easily separate and concentrate one titanium compound from the other) in remarkably short processing times was successfully tested with surrogate materials (TiO_2 and TiO) with different particle sizes, hardness and densities.

The major advantages of the RM-2 process are: fast grinding time, simple design and optimized design. These features make it significantly less costly than utilizing large-scale aqueous processes for disposal of small amounts of fissile material. This process may be able to provide a treatment that would reduce transportation costs, reduce the demand on safe and secure transportation equipment and reduce security cost for down blended uranium oxides.

In this research project the RM-2 milling technology was investigated using mixtures of natural uranium (NU) oxide, depleted uranium (DU) oxide, and ThO_2 as surrogates for HEU and U-233 materials to determine its capability to down-blend these materials.

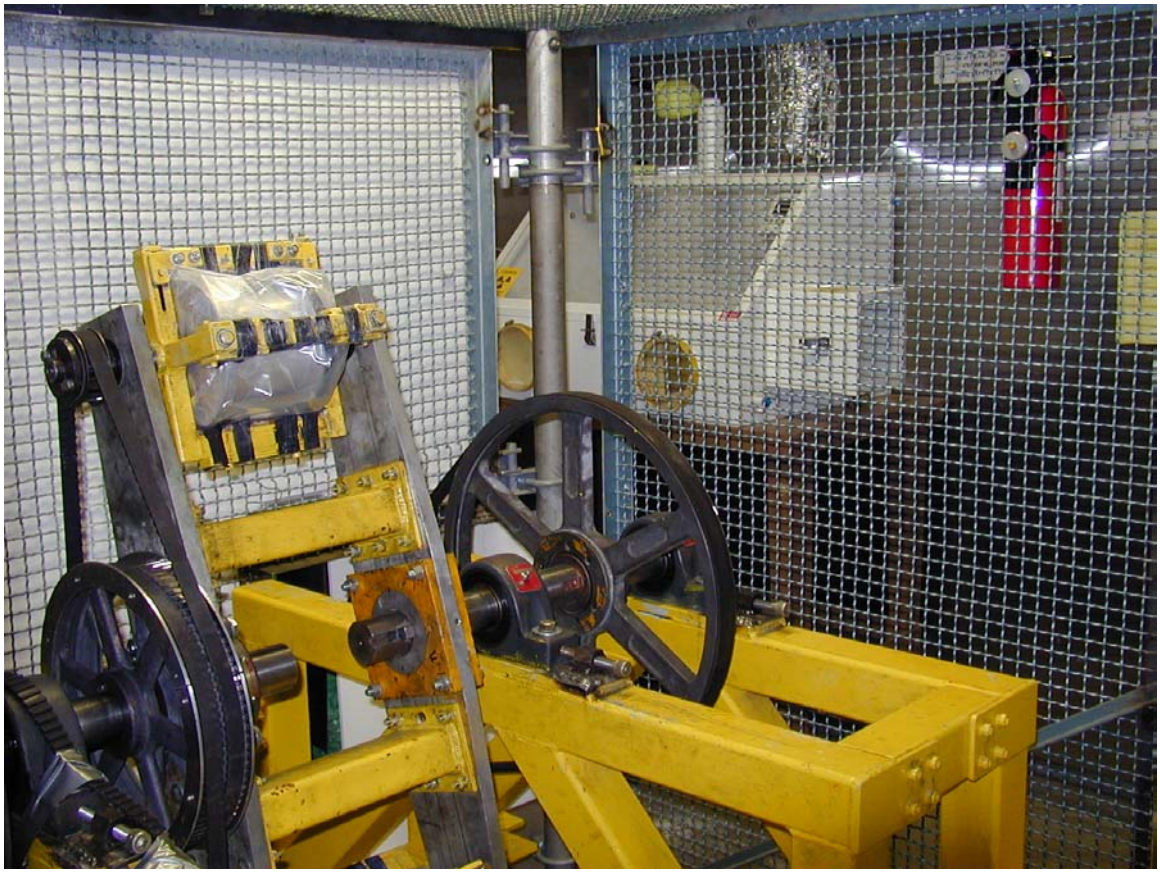


Figure 1. RM-2 Mill With Grinding Canister

WORK DESCRIPTION

These studies were scoped to demonstrate the RM-2 milling technology by testing with uranium oxide materials (DU and NU). The effects of mill operating and design variables on the grinding and blending of NU/DU oxides were studied with the intent of developing operating ranges for the process. Upon completion of these studies, the process parameters were used to outline a conceptual layout of a pilot scale facility to isotopically dilute HEU.

Three sets of tests were completed under this study.

- Milling mixtures of NU and DU of the same oxide (UO_3)
- Milling mixtures of NU and DU of mixed oxides (UO_2 with UO_3)
- Milling mixtures of LWBR pellets (ThO_2 surrogate with uranium oxide)

BLENDING TESTS FOR HIGHLY-ENRICHED U-235 SURROGATES

Two series of blending tests were performed in accordance with the approved management plan. In one series of tests, NU in the form of UO_3 was blended with DU in the form of UO_3. In the second series of test, NU in the form of UO_3 was blended with DU in the form of UO_2. The results of these tests are given in the following sections.

Blending Tests With Same Oxide Samples

A total of 12 grinding/blending tests have been completed using NU (in the form of UO_3) and DU (in the form of UO_3) primarily to understand the effects of grinding media balls (2 and 3 mm), void filling (50 and 75%), and grinding time (30, 60, and 90 minutes) on irreversible blending and product size distribution.

The products were analyzed by a VG Elemental PQ3 inductively-coupled plasma mass spectrometer (ICP-MS) at Argonne National Laboratory-West to determine the U-235/U-238 isotopic distribution. The particle size analyses of the products were conducted at ACTLab Canada. Figure 2 shows a typical particle size distribution for one set of test conditions.

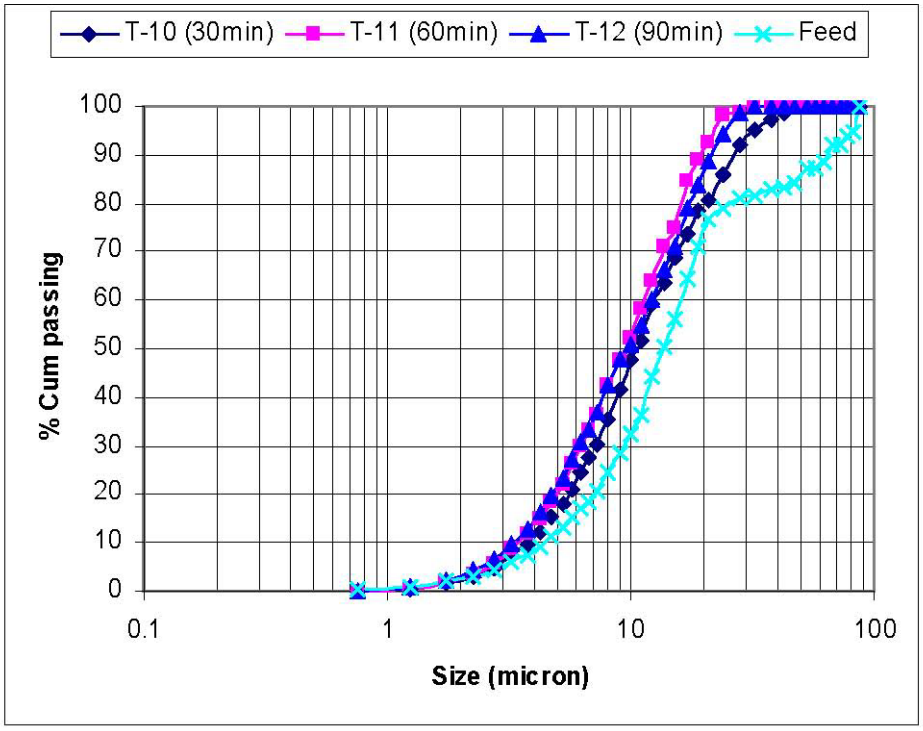


Figure 2. Size Distribution of Products at Different Grinding Times (2mm balls 70% Critical Speed)

The following observations have been made from these tests in regard to grinding and blending of the materials:

- The d50 (the size at which 50% of the mass passes through a specified sieve size) of the product is reached around 8 microns.
- Grinding with 2 mm balls gives finer and narrow size product compared to the 3 mm balls. This is due to more ball-to-particle collisions with 2 mm balls compared to the 3 mm balls. In addition the pore size (interstitial space) with 3 mm balls being larger, the particles can escape the collisions among balls.
- When the void filling increased from 50% to 75%, the product size distribution becomes much coarser. This is due to crowding of particles that reduces the ball-to-particle contact.
- When the mill critical speed was increased to 70% from 63%, the product becomes finer in relatively less grind time.
- With a progressive increase in grinding time, generally the product gets finer. In some instances, excessive grinding can result in negative grinding or agglomeration of ground particles.

Spatial samples of the ground product were taken and analyzed to determine the completeness of blending. In all tests the blended product fell within the band of standard error that was calculated indicating complete blending. The results are presented in Figure 3.

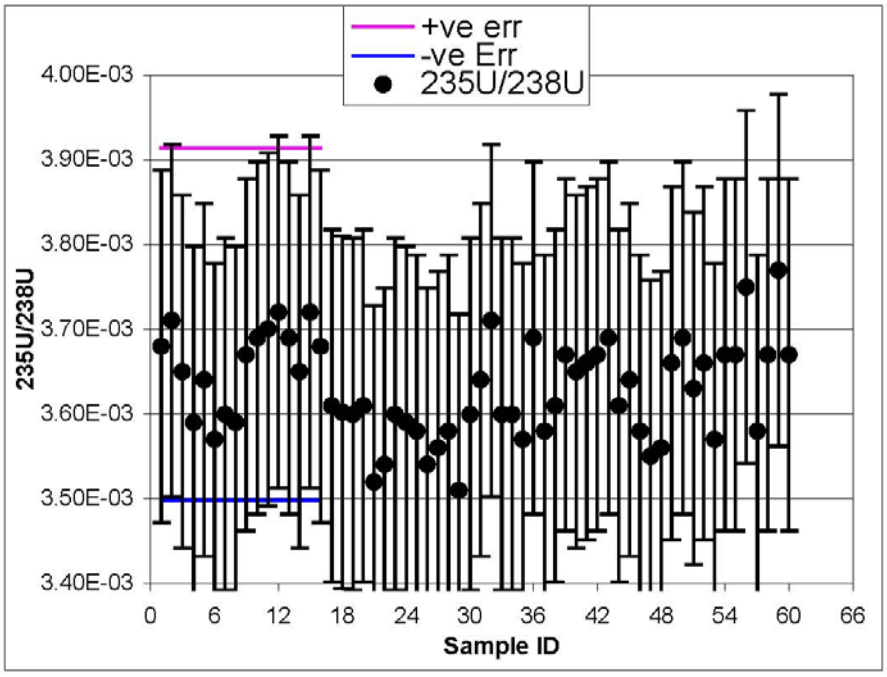


Figure 3. Isotopic Analysis of the Blended Product Shown Within the Band of Standard Error of Analysis

Blending Tests With Mixed Oxide Samples

A total of 22 grinding/blending tests have been completed with mixed oxide samples of NU (in the UO_3 form) and DU (DU in the UO_2 form) using four process variables. These variables are: grinding times of 30, 60, 90, and 150 minutes, grinding balls of 2 mm and 3 mm diameter, void filling of 50% and 75%, and critical mill speed of 75% and 100%. A ratio of 20:80 for NU+DU has been standard for these tests.

The products were analyzed by a VG Elemental PQ3 inductively-coupled plasma mass spectrometer (ICP-MS) at Argonne National Laboratory-West to determine the U-235/U-238 isotopic distribution. The particle size analyses of the products were conducted at ACTLab Canada. Figure 4 shows a typical particle size distribution for one set of test conditions.

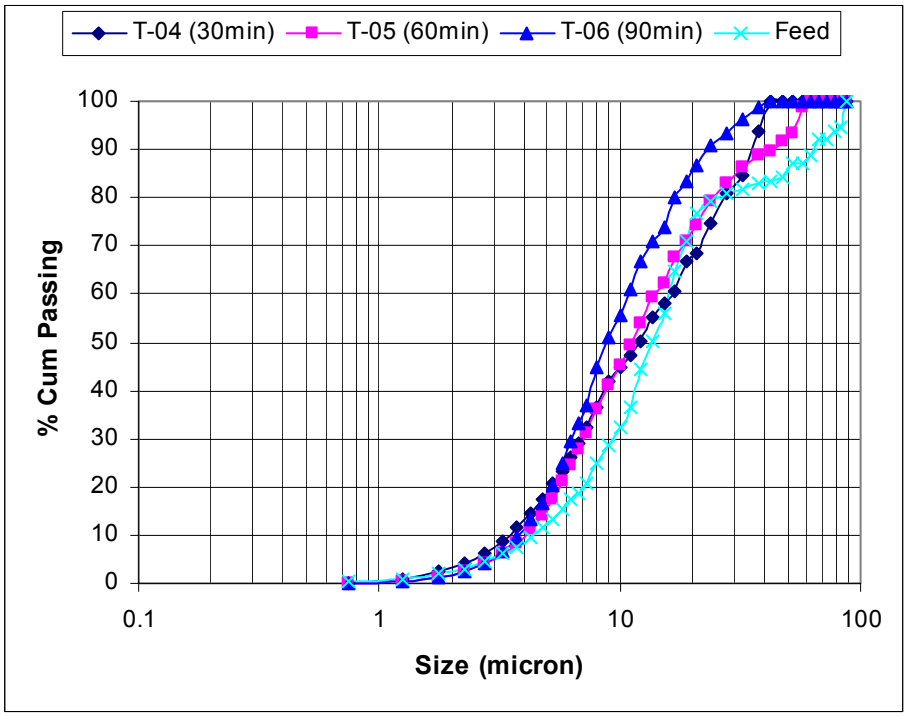


Figure 4. Size Distribution of Products at Different Grinding Times (2mm balls, 50% void filling, 100% critical mill speed)

The following observations have been made from these tests in regard to grinding and blending of the materials:

- The d50 of the product reached around 8 microns. This is readily achieved even at the shortest grinding/blending time studied.
- Grinding with 2 mm balls gives finer and narrow size product compared to the 3 mm balls. This is due to more ball-to-particle collisions with 2 mm balls compared to the 3 mm balls. In addition the pore size with 3 mm balls being larger, the particles can escape the collisions among balls.

- With increased grinding time, generally the product gets finer. In some instances, excessive grinding can result in negative grinding or agglomeration of particles.
- The distribution of U-235 and U-238 is constant throughout the volume of the blended material. This consistency occurs even at the shortest grinding/blending time.

From each final product batch, six spatial samples were taken and analyzed to determine the ratio of U-235 and U-238 throughout the product. This testing was needed to demonstrate that U-235 was not concentrated in any location of the milling canister. The results indicate that the blending of both NU and DU meets project expectations. The results fall within the standard errors of analysis suggesting that the blending is homogeneous. The results are presented in Figure 5.

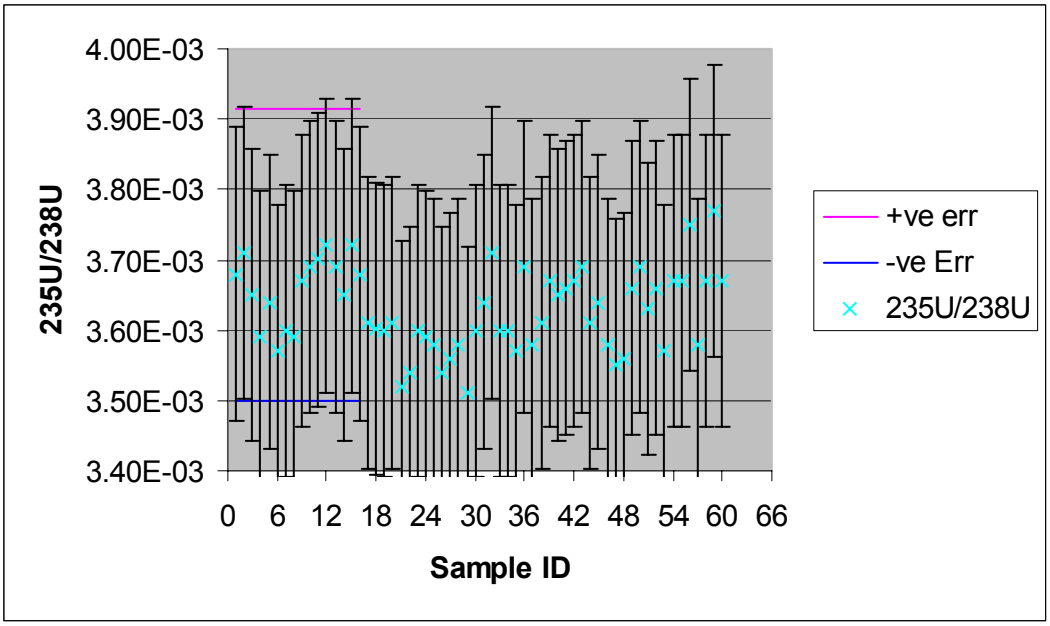


Figure 5. Isotopic Analysis of the Blended Product Shown Within the Band of Standard Error of ICP-MS Analysis

BLENDING TESTS FOR LWBR U-233 SURROGATES

The LWBR fuel pellets are a high-fired ceramic of uranium oxide (UO_2) in a predominately thorium oxide (ThO_2) matrix. The characteristics of this material provide separate and unique containment features for retaining U-233 via pellet integrity or toughness. Typically this oxide is very resistant to physical degradation because of its near massive crystalline structure that has very few crystalline defects. The surface of the pellet is very hard. Even if the hard ceramic pellet is physically broken the uranium will remain contained in the resulting pieces.

This study was conducted in two parts to simulate the steps that might be required to grind and blend LWBR fuel pellets to achieve isotopic dilution. This first part entailed the size reduction and blending of ThO_2 particles (325 mesh obtained from Alfa Assar

Inc.) with NU oxide particle to investigate the ability of the RM-2 mill to achieve blending and dilution. The second part of the study investigated the feasibility of size-reducing ceramic, high-fired ThO_2 pellets (obtained from Pacific Northwest National Laboratory) in the mill. The sequential approach was required because the schedule to obtain surrogate ThO_2-UO_2 pellets was beyond the period of performance for this study.

Eighteen blending tests with ThO_2 powder and NU feedstock were conducted while varying the process parameters of grinding ball size, void filling, critical speed, and time. Statistical analysis of the data implies that the entire mixture in the canister was completely mixed in all tests.

A particle size distribution was obtained from each test. Comparison of the initial ThO_2 and NU materials with the size distribution data from each test suggests that grinding times as low as twenty minutes is sufficient to obtain homogeneous blending and to obtain a uniform product size distribution. Figure 6 shows a typical size distribution obtained from these tests.

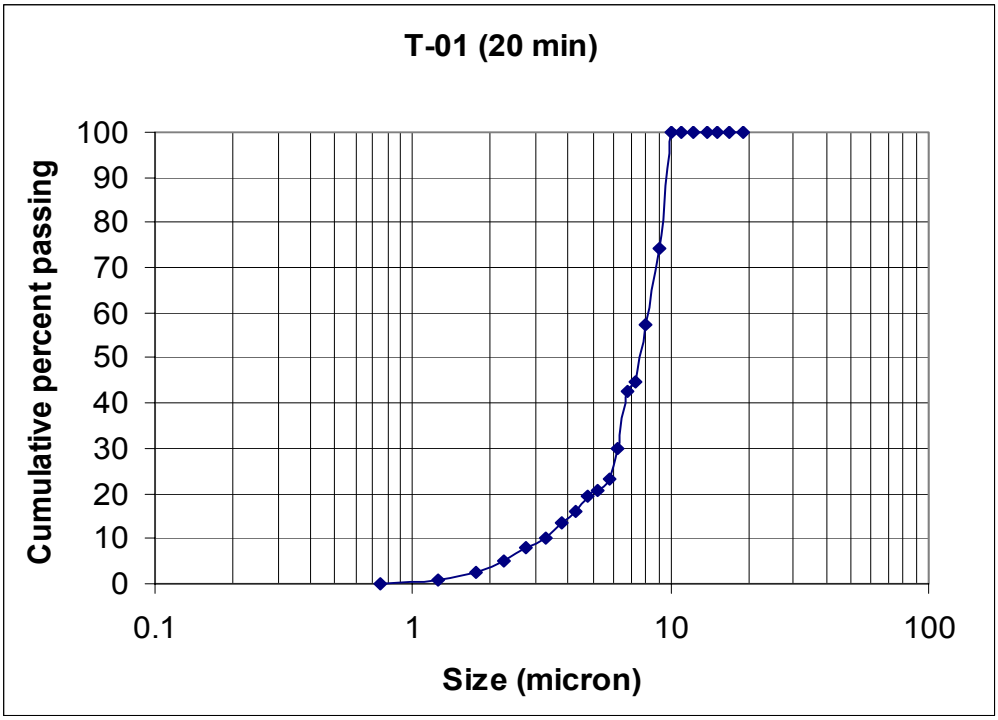


Figure 6. Product Size Distribution of Test-01C

The ability of the RM-2 mill to reduce surrogate LWBR fuel pellets was demonstrated in three tests using ThO_2 pellets with the approximate dimensions of 10 mm diameter and 6 mm in length. Magnesium oxide grinding balls were used while holding the process parameters of grinding ball fill, void fill, and speed constant and varying time. Figure 7 shows the ThO_2 pellets.

Figure 7. Thorium Oxide Pellets Received From PNNL.

It was observed that twenty minutes was sufficient to break all of the pellets to 100-micron size. As shown in Figure 8, after a grinding time of 60 minutes the size distribution obtained was nearly identical to the size distribution of the ThO_2 powder used in the blending tests.

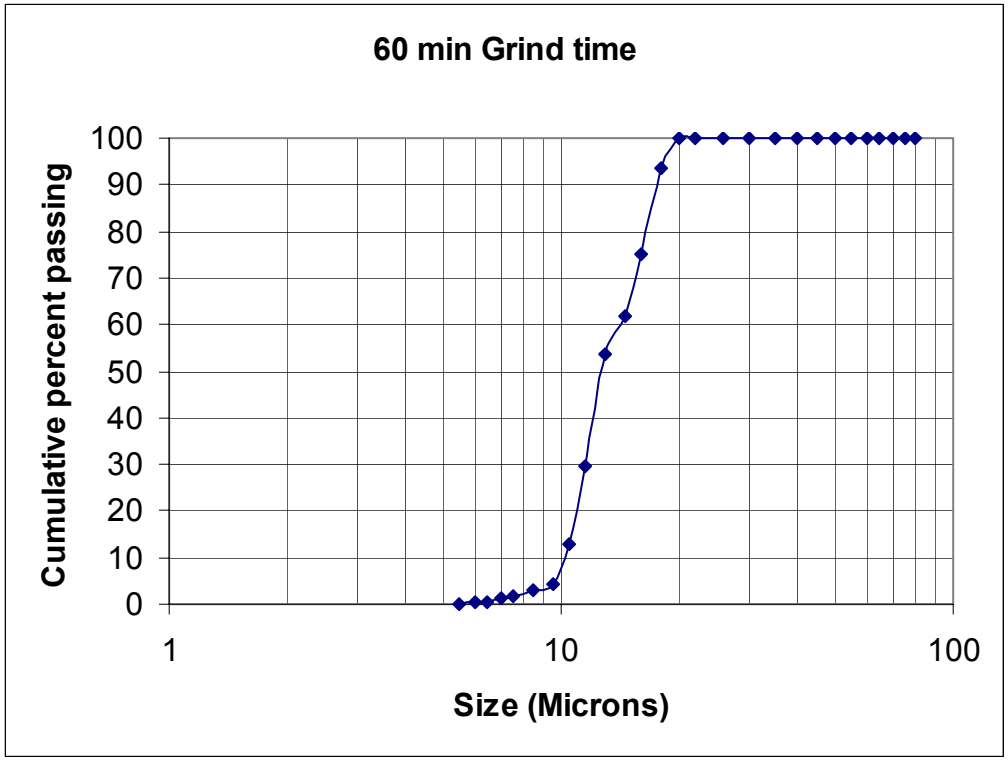


Figure 8. 60-Minute Grinding Product Size Distribution of ThO_2 Pellets

The results of these ThO_2 pellet and powder studies indicate that LWBR fuel pellets may be reduced in size and diluted with NU/DU to obtain isotopic dilution by the RM-2 mill

technology. Additional equipment (such as a crusher) may not be necessary to reduce the size of the LWBR pellets prior to grinding and blending in the RM-2 mill. However, such a step would potentially reduce the operational time.

CONCEPTUAL LAYOUT OF PILOT FACILITY

Based on the testing analytical data from these studies, a conceptual layout of a larger scale (8 kg of HEU as UO_3 per day or 40 kg per day total UO_3) RM-2 mill for an INEEL pilot facility was developed. The design specifications included, but were not necessarily limited to: grinding media removal, radiation shielding, safety requirement, product temperature control, remote handling, product packaging, grinding media packaging, exposure and infrastructure support requirements, mill and canister construction, sampling and non-destructive analysis techniques, and abnormal operating concerns and responses.

The conceptual layout of a pilot facility considered the dilution HEU stored at INTEC but did not consider the dilution of LWBR unirradiated fuel. The footprint of the pilot facility is constrained to be compatible with the area located in the vicinity of the north vault of CPP-651 at INTEC. The space available for the facility is an area of approximately 52 ft by 20 ft.

It was presumed that overhead cranes, ceiling exhaust, and lighting and other balance of plant equipment would be provided for the pilot facility. The floor area would be divided into three principal areas: feed loading area, blending area and product handling area, each separated by walls, doors, walk ways etc. Areas for DU storage and personal protective equipment (PPE) are also shown. The general footprint is shown in Figure 9. The 3-dimensional view of the floor plan is shown in Figure 10. The three major areas with additional details are shown in this figure.

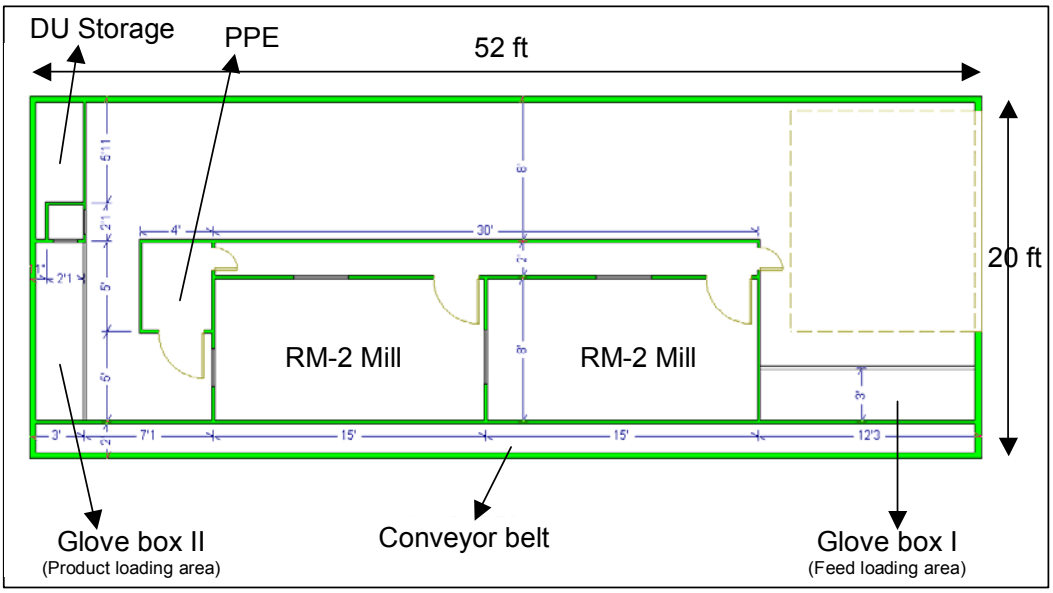


Figure 9. General Footprint of the Pilot Mill Facility at INEEL

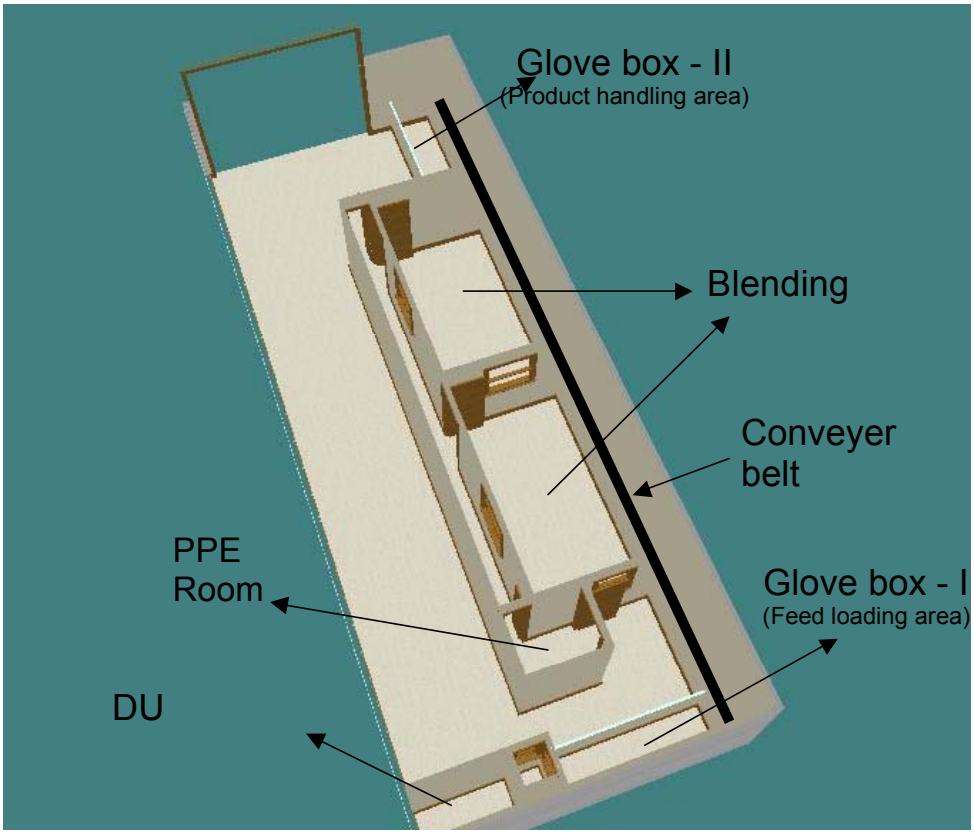


Figure 10. Three-Dimension View of the Pilot Mill Facility

RESULTS AND CONCLUSIONS

The results of this study indicate the RM-2 mill technology is applicable to the isotopic dilution of "off-spec" HEU located at the INEEL. Isotopic dilution could be accomplished for either recycle (dilute to <20 wt%) or disposal (dilute to 0.9 wt%) of this material.

Although more difficult to accomplish, the RM-2 technology could also be applied to the isotopic dilution of U-233 contained in LWBR unirradiated fuel. Additional size reduction equipment may be required before final milling and blending of materials by the RM-2 mill to achieve the desired isotopic dilution.

A pilot scale facility layout was developed to fit within an available area in the HEU storage facility located at INEEL. Although a larger footprint could provide an improved equipment and materials handling layout, major construction costs would be incurred to modify the existing facility. The layout within the existing facility appears to provide marginal operating space. Additional studies would be required to support a conceptual design.

ACKNOWLEDGEMENT

This work was a joint effort between the Idaho National Engineering and Environmental Laboratory and the University of Utah. Financial support provided by the National Energy Technology Laboratory under Solicitation DE-AC-01NT41312 is acknowledged.

BIBLOGRAPHY

R. Harmon, C. Vitale-Smith, R. N. Henry, "Dry Milling and Blending of High-Enriched Uranium Tri-Oxide," presented at American Nuclear Society-Department of Energy Spent Nuclear Fuel and Fissile Material Management Conference, Charleston SC, September, 1998.

R. N. Henry, R. J. Ramer, M. M. Plum, C. R. Kenley, "Engineering Study of the Disposition of Uranium-233 at the Idaho National Engineering and Environmental Laboratory," INEEL/EXT-2000-01588, January 2001.

R. N. Henry, N. A. Chipman, R. K. Rajamani, "Dry Blending to Achieve Isotopic Dilution of Highly Enriched Uranium Oxide Materials," presented at American Institute of Chemical Engineers Spring National Meeting, Houston TX, April, 2001.

M. K. Abdl El-Rehman, N. Ma, R. K. Rajamani, "Ultra-fine Grinding of Some Oxides and Non Oxides Materials Using the Planetary Mill," Ore Dressing/Cevher Hazirlama Journal, (5)2001, 11-23.

R. N. Henry, N. A. Chipman, R. K. Rajamani, J. L. Malhotra, "Nuclear Isotopic Dilution of Highly Enriched Uranium by Dry Blending," presented at Spectrum 2002, Reno NV, August 2002.

R. N. Henry, N. A. Chipman, R. K. Rajamani, S. Latchireddi, V. Devrani, H. Sethi, J. L. Malhotra, "Nuclear Isotopic Dilution of Highly Enriched Uranium-235 and Uranium-233 by Dry Blending Via the RM-2 Mill Technology," INEEL/EXT-03-01119, September 2003.